Inner-shell photodetachment of atomic and molecular negative ions

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INTRODUCTION

The importance of atomic and molecular negative ions has long been recognized in fields such as fundamental physics, atmospheric physics, interstellar chemistry, accelerator mass spectrometry, and others [1]. From a fundamental standpoint, photoexcitation and photodetachment processes in negative ions are a very sensitive probe of electron-correlation effects, and offer an important test for sophisticated theoretical models. While atomic and molecular negative ions have been studied extensively over the past three decades, investigations have been limited to outer-shell studies. Third generation synchrotron sources have recently made inner-shell photodetachment of negative ions readily achievable, with the first studies of inner-shell processes in atomic negative ions being performed less than two years ago [2 – 4]. Recent work by our group has concentrated on verifying the new theoretical results stimulated by those experiments, as well as performing the first inner-shell studies on molecular negative ions.

The present work was conducted with the ion-photon-beam end station on beamline 10.0.1 at the ALS. A Middleton type cesium sputter source (SNICS, NEC) and an RF rubidium charge-exchange ion source (Alphatross, NEC) are used to produce of a wide range of negative ion species with an ion beam energy of generally 7 – 10 keV. The ion beam is mass-analyzed in a magnetic field and electrostatically merged with the photon beam from beamline 10.0.1. Positive ions formed from photodetachment or photoexcitation of the negative ions, and subsequent Auger-decay, are detected as a function of photon energy. In the case of molecular negative ions, fragmentation of the parent ion from the core-excitation and detachment processes results in the production of charged atomic and molecular products which are mass-analyzed and detected separately. Possible neutral atomic and molecular products cannot be detected at the present time.

THE NEGATIVE ION OF HELIUM

Although He^- has only 3 electrons, it presents a significant challenge for theoretical calculations. Significant discrepancies with theoretical predictions were observed in the only previous experimental core-detachment study conducted on He^- to date [5]. In particular, some of the structure observed was inconsistent with theory and the magnitude of the double-detachment cross section near the He 2s2p $^3P^o$ threshold appeared to be smaller than expected. These experimental results have attracted considerable theoretical interest this past year [6,7]. The structure located about 4.5 eV above the 2s2p $^3P^o$ threshold, which was previously believed to be nonresonant, was reinvestigated with new *ab initio* calculations. These calculations have indicated that the structure should instead be ascribed to “hollow ion” triply-excited He quartet resonances. A comparison of the predicted and measured positions, widths, and excitation strengths of these resonances provide a strong test for these new theoretical investigations.

Preliminary results of recent high-resolution measurements of the triply-excited He^- quartet states are presented in Figure 1. The recent theoretical results appear in good agreement with these new measurements. In addition, a third resonance (not shown in Figure 1), was observed in the He^+ product channel about 0.9 eV below the 2s2p $^3P^o$ threshold, and identified as arising from the He^- $2s2p^2$ $^4P^e$ state. Since there are no intermediate states present for sequential decay of this resonance through a neutral atomic state, this represents the first observation of a simultaneous three-electron decay process from a photoexcited negative ion. Finally, direct measurements of the absolute double-photodetachment cross section near the 2s2p $^3P^o$ threshold (located 38.6 eV above the He^- 1s2s2p $^4P^o$ ground state) were also conducted.

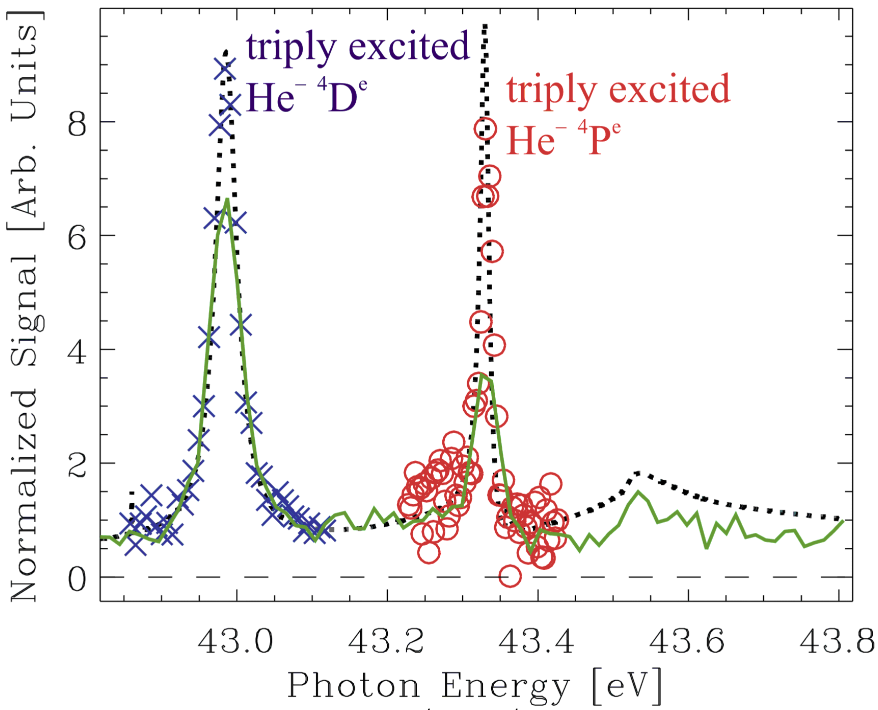

Figure 1. He^+ photoion yield from the triply excited He^- $^4D^e$ and $^4P^e$ states. The measurements were performed at three different spectral resolutions: 40 meV (solid curve), 20 meV (×), and 10 meV (○). Theory [5] is shown with a dotted curve, scaled to the observed signal. For easy comparison, the photon energy has been shifted to match the theory at the $^4D^e$ resonance; direct calibration of the experimental photon-energy is in progress.

NEGATIVE ION CLUSTERS

Figure 2 shows preliminary results of the first studies of inner-shell detachment of negative ion clusters. These investigations focused on K-shell detachment measurements of B^-, B_2^-, and B_3^-. All singly-charged positive ion fragments were observed. No detectable signal was observed in the negative ion and multiply-charged positive ion fragment channels. The results show clear energy shifts and amplitude changes of the near-threshold structure, arising at least in part from the B^- $1s\,2s^2\,2p^3$ ^3D, ^3S, and ^3P states. At higher spectral resolutions, the strongest peak in both the B^- and B_2^- spectra was resolved into two resonances. Analysis of the measured absolute cross sections for positive ion formation and the fragmentation product channel strength ratios (branching ratios) is underway. Similar work has also been conducted in L-shell detachment of silicon negative ion atom and clusters (Si^-, Si_2^-, and Si_3^-). That work covered an 80 eV photon energy range near the 2p and 2s thresholds, and showed no prominent resonant structure.

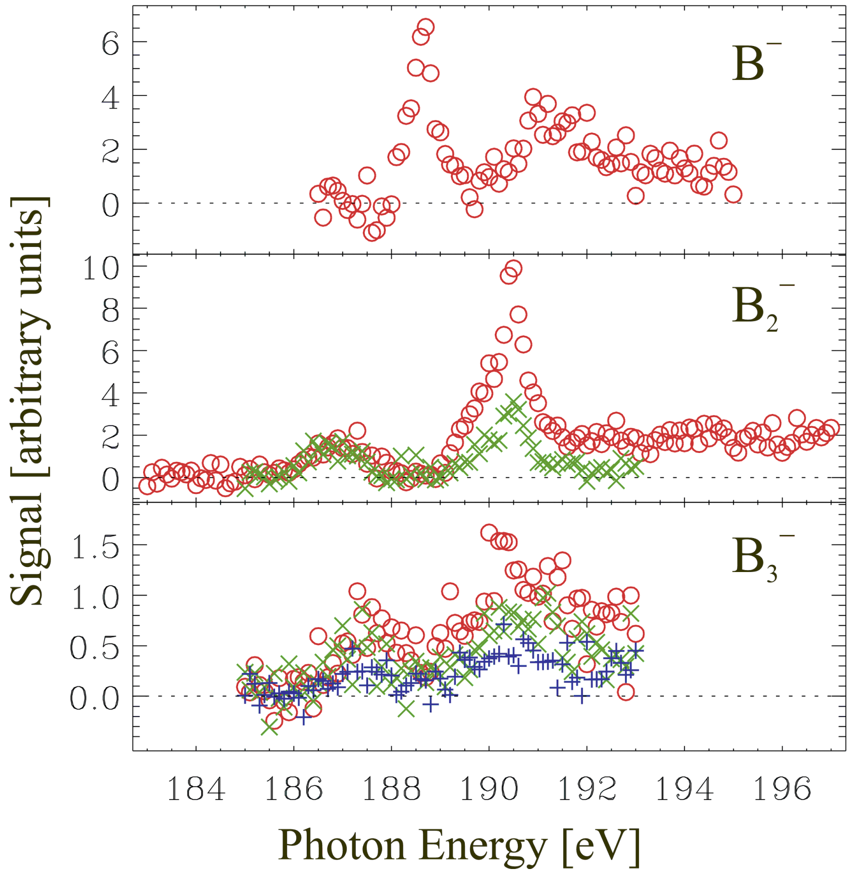

Figure 2. B^+ (○), B_2^+ (×), and B_3^+ (+) ion yields from the core photodetachment and photoexcitation of B^- (upper panel), B_2^- (middle panel), and B_3^- (lower panel). This data was taken at a 400 meV spectral resolution.

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